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MAR 31 2006

**Certificate**

APR 04 2006

**of Correction**

<b>To:</b>	<b>USPTO – Mail Certificate of Correction Branch</b>	<b>From:</b>	<b>Jason D. Voight</b>
<b>Fax:</b>	<b>(571) 273-8300</b>	<b>Pages:</b>	<b>6</b>
<b>Phone:</b>	<b>(571) 272-1000</b>	<b>Date:</b>	<b>March 31, 2006</b>
<b>Re:</b>	<b>Attorney Docket: 50512</b>	<b>cc:</b>	

☐ Urgent    ☐ For Review    ☐ Please Comment    ☐ Please Reply    ☐ Please Recycle

• **Comments:**

In re Application of: **MÜLLER et al.**  
Patent No.: **6,867,326**  
Issued: **March 15, 2005**  
Serial No.: **10/031,234**  
Filing Date: **July 21, 2000**  
Title: **METHOD OF PRODUCING GLYPHOSPHATE OR A SALT THEREOF**  
Attachments: **Request for Certificate of Correction  
Form PTO/SB44  
Mark-up copy of Patent**

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## IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re the patent of ) Certificate of Corrections Branch  
)  
MÜLLER et al. )  
)  
Patent No. 6,867,326 )  
)  
Date of Patent: 15 March 2005 )

For: METHOD OF PRODUCING GLYPHOSPHATE OR A SALT THEREOF

I hereby certify that this correspondence is being deposited with the United States Postal Service with sufficient postage as first class mail in an envelope addressed to Commissioner for Patents, P.O. Box 1450, Alexandria, Virginia 22313-1450, or being facsimile transmitted to the United States Patent and Trademark Office, Fax No. 571-273-8300, on 3/31/06.

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*Michelle Bryant*

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REQUEST FOR CERTIFICATE OF CORRECTION UNDER 37 CFR 1.322

Applicants request a Certificate of Correction for this patent. A form PTO/SB/44 is enclosed.

Because these mistakes were incurred through the fault of the Office, no fee should be due.

Please charge any shortage in fees due in connection with the filing of this paper to Deposit Account No. 14.1437. Please credit any excess fees to such account.

Respectfully submitted,  
NOVAK DRUCE DeLUCA & QUIGG LLP

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PTO/SB/44 (04-05)

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## UNITED STATES PATENT AND TRADEMARK OFFICE CERTIFICATE OF CORRECTION

Page 1 of 2

PATENT NO. : 6,867,326

APPLICATION NO.: 10/031,324

ISSUE DATE : 03/15/2005

INVENTOR(S) : MÖLLER et al.

It is certified that an error appears or errors appear in the above-identified patent and that said Letters Patent is hereby corrected as shown below:

In Claim 5, column 8, line 38 please delete:

"COO, HEU, IFR, ISV, ITE, JBW, KFI, LAU, LEV, LIO," and substitute therefore:

– GOO, HEU, IFR, ISV, ITE, JBW, KFI, LAU, LEV, LIO, –

In Claim 8, column 9, line 25 please delete:

"MWW, NAT, NES, NON, OFF, OSI PAR, PAU, PHI, RHO," and substitute therefore:

– MWW, NAT, NES, NON, OFF, OSI, PAR, PAU, PHI, RHO, –

In Claim 9, column 9, line 33 please delete:

"IIa, IVa, Va, VIIa, VIIa, VIIa, Ib, IIb, IIb, IVb, Vb, VIIb," and substitute therefore:

– IIIa, IVa, Va, VIa, VIIa, VIIa, Ib, IIb, IIb, IVb, Vb, VIIb, –

In Claim 10, column 9, line 37 please delete:

"element selected from among the elements of groups Ia, Ia," and substitute therefore

– element selected from among the elements of groups Ia, IIa, –

In Claim 10, column 9, line 38 please delete:

"IIa, IVa, Va, VIIa, VIIa, VIIa, Ib, IIb, IIb, IVb, Vb, VIIb," and substitute therefore:

– IIIa, IVa, Va, VIa, VIIa, VIIa, Ib, IIb, IIb, IVb, Vb, VIIb, –

In Claim 11, column 9, line 43 please delete:

"IIa, IVa, Va, VIIa, VIIa, VIIa, Ib, IIb, IIb, IVb, Vb, VIIb," and substitute therefore:

– IIIa, IVa, Va, VIa, VIIa, VIIa, Ib, IIb, IIb, IVb, Vb, VIIb, –

In Claim 12, column 9, line 48 please delete:

"IIa, IVa, Va, VIIa, VIIa, VIIa, Ib, IIb, IIb, IVb, Vb, VIIb," and substitute therefore:

– IIIa, IVa, Va, VIa, VIIa, VIIa, Ib, IIb, IIb, IVb, Vb, VIIb, –

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PTO/SB/44 (04-05)

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## UNITED STATES PATENT AND TRADEMARK OFFICE CERTIFICATE OF CORRECTION

Page 2 of 2

PATENT NO. : 6,867,326

APPLICATION NO.: 10/031,324

ISSUE DATE : 03/15/2005

INVENTOR(S) : MÖLLER et al.

It is certified that an error appears or errors appear in the above-identified patent and that said Letters Patent is hereby corrected as shown below:

In Claim 13, column 9, line 53 please delete:

"IIla, IVa, Va, VIIa, VIIa, VIIa, Ib, IIb, IIb, IVb, Vb, VIIb," and substitute therefore:

– IIIa, IVa, Va, VIa, VIIa, VIIa, Ib, IIb, IIb, IVb, Vb, VIIb, –

In Claim 21, column 10, line 57 please delete:

"IIla, IVa, Va, VIIa, VIIa, VIIa, Ib, IIb, IIb, IVb, Vb, Vab," and substitute therefore:

– IIIa, IVa, Va, VIa, VIIa, VIIa, Ib, IIb, IIb, IVb, Vb, VIIb, –

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(II) treating the catalyst at a temperature in the range from 250 to 800° C., preferably from 350 to 600° C., with a gas stream having a content of an oxygen-donating substance or of oxygen or of a mixture of two or more thereof in the range from 0.1 to 4% by volume, and optionally the further steps (II) and (IV),

(III) treating the catalyst at a temperature in the range from 250 to 800° C., preferably from 350 to 600° C., with a gas stream having a content of an oxygen-donating substance or of oxygen or of a mixture of two or more thereof in the range from >4 to 100% by volume,

(IV) cooling the regenerated catalyst obtained in step (III) in an inert gas stream containing up to 20% by volume of the vapor of a liquid selected from the group consisting of water, alcohols, aldehydes, ketones, ethers, acids, esters, nitrates, hydrocarbons and mixtures of two or more thereof.

Details of this process may be found in DE-A 197 23 949.8.

As regards the reaction conditions in the process of the present invention, there are no particular restrictions. The reaction can be carried out at subatmospheric, atmospheric or superatmospheric pressures, depending on the reaction temperature used. This is generally in the range from about 25 to about 150° C., preferably from about 50 to about 120° C. and in particular from about 70 to about 100° C.

As solvent, it is possible to use either organic solvents or water or mixtures thereof. Preference is given to carrying out the reaction in aqueous solution or suspension.

The reaction time is in the range from one minute to a plurality of hours. There are no restrictions in respect of the molar ratio of oxidant to PMIDE; preference is given to using at least 0.5 mol, more preferably at least 1 mol, of oxidant per mol of PMIDE.

As already mentioned, the reaction mixture can be brought into contact with the catalyst either in suspension or in a fixed bed. The reaction can be carried out batchwise or continuously.

To avoid safety risks, the oxygen content during the reaction is generally set so that the composition is reliably outside the explosive limits. This is achieved, for example, by mixing in suitable inert gases such as nitrogen.

The present invention is illustrated by the examples below:

## EXAMPLES

### Example 1

In a 250 ml three-necked flask fitted with stirrer, dropping funnel and reflux condenser, 20 g of catalyst (Cr/zeolite having a BEA structure and a chromium content of 2.8% by weight) in 100 ml of deionized water were combined with 34 g of PMIDE and heated to 85° C. while stirring. After this temperature had been reached, 70 ml of a 30% strength by weight H<sub>2</sub>O<sub>2</sub> solution were added and the solution was allowed to react for 1 hour. After cooling, the catalyst was separated off and the clear reaction solution was analyzed. The glyphosate content was determined as 3.2% by weight by means of HPLC using UV detection; this corresponds to a yield of 18 mol % based on PMIDE used.

### Example 2

Example 1 was repeated using 20 g of montmorillonite (bulk density: 670 g/l; specific surface area: 230 m<sup>2</sup>/g) as catalyst. The glyphosate content of the resulting reaction mixture was 6.60% by weight, corresponding to a yield of 44% based on PMIDE used.

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We claim:

1. A process for preparing N-phosphonomethylglycine or a salt thereof by bringing phosphonomethyliminodiacetic acid or a salt thereof into contact with at least one oxygen-containing oxidant in the presence of a heterogeneous catalyst comprising at least one silicate, with the proviso that the process is not an oxidative cleavage of N-phosphonomethyliminodiacetic acid with simultaneous oxidation of formaldehyde formed as by-product in the presence of a catalyst system which comprises activated carbon together with a microporous acid-resistant aluminosilicate having a ratio of Si to Al of at least 2 as support and a noble metal as active component.

2. A process as claimed in claim 1, wherein the oxygen-containing oxidant or oxidants is/are selected from the group consisting of hydroperoxides, gases containing molecular oxygen, oxygen-donating compounds, nitrogen oxides and mixtures of two or more thereof.

3. A process as claimed in claim 1 wherein the silicate or silicates is/are selected from the group consisting of zeolites, sheet silicates, naturally occurring or synthetically produced clay minerals, clathrasils and mixtures of two or more thereof.

4. A process as claimed in claim 2, wherein the silicate or silicates is/are selected from the group consisting of zeolites, sheet silicates, naturally occurring or synthetically produced clay minerals, clathrasils and mixtures of two or more thereof.

5. A process as claimed in claim 1, wherein the crystalline silicate or silicates is/are selected from the group consisting of zeolite structure-types having framework type codes ABW, ACO, AEL, AEL, AEN, AET, AFG, AFL, AFN, AFO, AFR, AFS, AFT, AFX, AFY, AHT, ANA, APC, APD, AST, ATN, ATO, ATS, ATT, ATV, AWO, AWW, BEA, BIK, BOG, BPH, BRE, CAN, CAS, CFL, CGF, CGS, CHA, CHI, CLO, CON, CZP, DAC, DDR, DFO, DFT, DOH, DON, EAB, EDI, EMT, EPI, ERI, ESV, EUO, FAU, FER, GIS, GME, GOO, HEU, IFR, ISV, ITE, JBW, KFI, LAU, LEV, LIO, LOS, LOV, LTA, LTL, LTN, MAZ, MEL, MEL, MEP, MER, MFI, MFS, MON, MOR, MSO, MTF, MTN, MTT, MTW, MWW, NAT, NES, NON, OFF, OSI, PAR, PAU, PHI, RHO, RON, RSN, RTE, RTH, RUT, SAO, SAT, SBE, SBS, SBT, SFF, SGT, SOD, STF, STL, STT, TER, THO, TON, TSC, VET, VFI, VNI, VSV, WEL, WEN, YUG and ZON, and mixtures of two or more thereof.

6. A process as claimed in claim 2, wherein the crystalline silicate or silicates is/are selected from the group consisting of zeolite-structure types having framework type codes ABW, ACO, AEL, AEL, AEN, AET, AFG, AFL, AFN, AFO, AFR, AFS, AFT, AFX, AFY, AHT, ANA, APC, APD, AST, ATN, ATO, ATS, ATT, ATV, AWO, AWW, BEA, BIK, BOG, BPH, BRE, CAN, CAS, CFL, CGF, CGS, CHA, CHI, CLO, CON, CZP, DAC, DDR, DFO, DFT, DOH, DON, EAB, EDI, EMT, EPI, ERI, ESV, EUO, FAU, FER, GIS, GME, GOO, HEU, IFR, ISV, ITE, JBW, KFI, LAU, LEV, LIO, LOS, LOV, LTA, LTL, LTN, MAZ, MEL, MEL, MEP, MER, MFI, MFS, MON, MOR, MSO, MTF, MTN, MTT, MTW, MWW, NAT, NES, NON, OFF, OSI, PAR, PAU, PHI, RHO, RON, RSN, RTE, RTH, RUT, SAO, SAT, SBE, SBS, SBT, SFF, SGT, SOD, STF, STL, STT, TER, THO, TON, TSC, VET, VFI, VNI, VSV, WEL, WEN, YUG and ZON, and mixtures of two or more thereof.

7. A process as claimed in claim 3, wherein the crystalline silicate or silicates is/are selected from the group consisting of zeolite-structure types having framework type codes ABW, ACO, AEL, AEL, AEN, AET, AFG, AFL, AFN, AFO, AFR, AFS, AFT, AFX, AFY, AHT, ANA, APC, APD, AST,

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ATN, ATO, ATS, ATT, ATV, AWO, AWW, BEA, BIK, BOG, BPH, BRE, CAN, CAS, CFI, CGF, CGS, CHA, CHI, CLO, CON, CZP, DAC, DDR, DFO, DFT, DOH, DON, EAB, EDI, EMT, EPI, ERI, ESV, EUO, FAU, FER, GIS, GME, GOO, HEU, IFR, ISV, ITE, JBW, KFI, LAU, LEV, LIO, LOS, LOV, LTA, LTL, LTN, MAZ, MEL, MEF, MER, MFI, MFS, MON, MOR, MSO, MTF, MTN, MTT, MTW, MWW, NAT, NES, NON, OFF, OSI, PAR, PAU, PHI, RHO, RON, RSN, RTE, RTH, RUT, SAO, SAT, SBE, SBS, SBT, SFF, SGT, SOD, STF, STL, TER, THO, TON, TSC, VET, VFI, VNI, VSV, WEI, WEN, YUG and ZON, and mixtures of two or more thereof.

8. A process as claimed in claim 4, wherein the crystalline dilute or silicates is/are selected from the group consisting of zeolite-structure types having framework type codes ABW, ACO, AEI, AEL, AEN, AET, AFG, AFL, APN, APO, AFR, AFS, AFT, AFX, AFY, AHT, ANA, APC, APD, AST, ATN, ATO, ATS, ATT, ATV, AWO, AWW, BEA, BIK, BOG, BPH, BRE, CAN, CAS, CFI, CGF, CGS, CHA, CHI, CLO, CON, CZP, DAC, DDR, DFO, DFT, DOH, DON, EAB, EDI, EMT, EPI, ERI, ESV, EUO, FAU, FER, GIS, GME, GOO, HEU, IFR, ISV, ITE, JBW, KFI, LAU, LEV, LIO, LOS, LOV, LTA, LTL, LTN, MAZ, MEL, MEF, MER, MFI, MFS, MON, MOR, MSO, MTF, MTN, MTT, MTW, MWW, NAT, NES, NON, OFF, OSI, PAR, PAU, PHI, RHO, RON, RSN, RTE, RTH, RUT, SAO, SAT, SBE, SBS, SBT, SFF, SGT, SOD, STF, STL, TER, THO, TON, TSC, VET, VFI, VNI, VSV, WEI, WEN, YUG and ZON, and mixtures of two or more thereof.

9. A process as claimed in claim 1, wherein the at least one heterogeneous catalyst further comprises at least one element selected from among the elements of groups Ia, IIa, IIIa, IVa, Va, VIa, VIIa, VIIIa, Ib, IIb, IIIb, IVb, Vb, VIb, VIIb of the Periodic Table.

10. A process as claimed in claim 2, wherein the at least one heterogeneous catalyst further comprises at least one element selected from among the elements of groups Ia, IIa, IIIa, IVa, Va, VIa, VIIa, VIIIa, Ib, IIb, IIIb, IVb, Vb, VIb, VIIb of the Periodic Table.

11. A process as claimed in claim 3, wherein the at least one heterogeneous catalyst further comprises at least one element selected from among the elements of groups Ia, IIa, IIIa, IVa, Va, VIa, VIIa, VIIIa, Ib, IIb, IIIb, IVb, Vb, VIb, VIIb of the Periodic Table.

12. A process as claimed in claim 4, wherein the at least one heterogeneous catalyst further comprises at least one element selected from among the elements of groups Ia, IIa, IIIa, IVa, Va, VIa, VIIa, VIIIa, Ib, IIb, IIIb, IVb, Vb, VIb, VIIb of the Periodic Table.

13. A process as claimed in claim 5, wherein the at least one heterogeneous catalyst further comprises at least one element selected from among the elements of groups Ia, IIa, IIIa, IVa, Va, VIa, VIIa, VIIIa, Ib, IIb, IIIb, IVb, Vb, VIb, VIIb of the Periodic Table.

14. A process as claimed in claim 1, wherein the heterogeneous catalyst is regenerated after complete or partial loss of activity and the regenerated heterogeneous catalyst is reused for preparing N-phosphonomethylglycine or a salt thereof from phosphonomethyliminodiacetic acid or a salt thereof.

15. A process as claimed in claim 2, wherein the heterogeneous catalyst is regenerated after complete or partial loss of activity and the regenerated heterogeneous catalyst is reused for preparing N-phosphonomethylglycine or a salt thereof from phosphonomethyliminodiacetic acid or a salt thereof.

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16. A process as claimed in claim 3, wherein the heterogeneous catalyst is regenerated after complete or partial loss of activity and the regenerated heterogeneous catalyst is reused for preparing N-phosphonomethylglycine or a salt thereof from phosphonomethyliminodiacetic acid or a salt thereof.

17. A process as claimed in claim 4, wherein the heterogeneous catalyst is regenerated after complete or partial loss of activity and the regenerated heterogeneous catalyst is reused for preparing N-phosphonomethylglycine or a salt thereof from phosphonomethyliminodiacetic acid or a salt thereof.

18. A process as claimed in claim 5, wherein the heterogeneous catalyst is regenerated after complete or partial loss of activity and the regenerated heterogeneous catalyst is reused for preparing N-phosphonomethylglycine or a salt thereof from phosphonomethyliminodiacetic acid or a salt thereof.

19. A process for preparing N-phosphonomethylglycine or a salt thereof by bringing phosphonomethyliminodiacetic acid or a salt thereof into contact with at least one oxygen-containing oxidant selected from a group consisting of hydroperoxides, gases containing molecular oxygen, oxygen-donating compounds, nitrogen oxides and mixtures of two or more thereof, in the presence of a heterogeneous catalyst, comprising at least one silicate selected from the group consisting of zeolites, sheet silicates, naturally occurring or synthetically produced clay minerals, clays and mixtures of two or more thereof, with the proviso that the process is not an oxidative cleavage of N-phosphonomethyliminodiacetic acid with simultaneous oxidation of formaldehyde formed as by-product in the presence of a catalyst system which comprises activated carbon together with a microporous acid-resistant aluminosilicate having a ratio of Si to Al of at least 2 as support and a noble metal as active component.

20. A process as claimed in claim 19, wherein the zeolites are selected from the group consisting of zeolites of the structure types having framework type codes ABW, ACO, AEI, AEL, AEN, AET, AFG, AFL, APN, APO, AFR, AFS, AFT, AFX, AFY, AHT, ANA, APC, APD, AST, ATN, ATO, ATS, ATT, ATV, AWO, AWW, BEA, BIK, BOG, BPH, BRE, CAN, CAS, CFI, CGF, CGS, CHA, CHI, CLO, CON, CZP, DAC, DDR, DFO, DFT, DOH, DON, EAB, EDI, EMT, EPI, ERI, ESV, EUO, FAU, FER, GIS, GME, GOO, HEU, IFR, ISV, ITE, JBW, KFI, LAU, LEV, LIO, LOS, LOV, LTA, LTL, LTN, MAZ, MEL, MEF, MEF, MER, MFI, MFS, MON, MOR, MSO, MTF, MTN, MTT, MTW, MWW, NAT, NES, NON, OFF, OSI, PAR, PAU, PHI, RHO, RON, RSN, RTE, RTH, RUT, SAO, SAT, SBE, SBS, SBT, SFF, SGT, SOD, STF, STL, TER, THO, TON, TSC, VET, VFI, VNI, VSV, WEI, WEN, YUG and ZON, and mixtures of two or more thereof.

21. A process as claimed in claim 19, wherein the at least one heterogeneous catalyst further comprises at least one element selected from among the elements of groups Ia, IIa, IIIa, IVa, Va, VIa, VIIa, VIIIa, Ib, IIb, IIIb, IVb, Vb, VIb, VIIb of the Periodic Table.

22. A process as claimed in claim 19, wherein the heterogeneous catalyst is regenerated after complete or partial loss of activity and the regenerated heterogeneous catalyst is reused for preparing N-phosphonomethylglycine or a salt thereof from phosphonomethyliminodiacetic acid or a salt thereof.

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